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Selenium Transport and Bioaccumulation in Aquatic Ecosystems: A Proposal for Water Quality Criteria Based on Hydrological Units

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Local water quality criteria for selenium should be based on an assessment of the degree of toxicological hazard to fish and wildlife, which is influenced by the spatial and temporal variation of the selenium cycle at the site under consideration. The physical area from which measurements are taken to evaluate selenium residues and biological effects, i.e., the database for setting site-specific criteria, must encompass more than an isolated segment of river, a tributary stream, etc. Because of hydrological connections between the various aquatic habitats that may be present in a watershed basin—wetlands, rivers, streams, and impoundments—the toxic threat from selenium contamination is also connected. For example, a criterion that is appropriate for a stream or river where low bioaccumulation occurs may result in *seemingly* harmless concentrations of selenium becoming a problem in downstream impoundments or in off-channel bays and wetlands where bioaccumulation is greater. The hydrologically connected parts of a basin downstream of a selenium discharge (natural or synthetic selenium source), extending to the point at which new sources of low-selenium water dominate the hydrology (e.g., confluence with larger tributary or river, spring or groundwater inflow), should be the area evaluated and given a specific criterion, not isolated components. Thus, a hydrological unit should be identified and used as the "site" for the purpose of setting criteria. Importantly, criteria derived in such a fashion will reflect the transport and bioaccumulation of selenium within the entire hydrological unit rather than simply focusing on a small, artificially designated segment of the system. Failure to use a hydrological unit approach can set the stage for significant biological and legal problems.

INTRODUCTION

The processes that regulate how selenium moves through the components of an aquatic habitat, as well as the presence of different habitat types within many watershed basins (wetlands, streams, rivers, impoundments), have important implications for setting water quality criteria for this trace element. In the decade since the U.S. Environmental Protection Agency (EPA, Agency) established the current national criteria (USEPA, 1987) there have been several field-documented cases of toxic effects to wetland biota when waterborne concentrations of selenium were below the criterion value for chronic exposure (5 μg/liter), as well as observations that suggest that the criterion is too restrictive for some rivers and streams (e.g., Canton and Van Derveer, 1997; Van Derveer and Canton, 1997; Hallock and Hallock, 1993; Hamilton *et al.*, 1996; Lemly, 1995a, Stephens *et al.*, 1992; Skorupa, 1998; Skorupa and Ohlendorf, 1991). This has led several states, the U.S. Fish and Wildlife Service, and local municipalities to pursue development and implementation of criteria on a site-specific basis (e.g., Guglielmone, 1995). Although this is a legitimate course of action, the approaches used to derive criteria are often seriously flawed because certain components of the selenium cycle are overlooked or underestimated, especially the potential for bioaccumulation and toxic impacts downstream of the "site" under consideration (Hamilton and Lemly, in press). Moreover, the motivation of some who seek to modify criteria may be to escape requirements for reducing selenium discharges rather than to establish criteria based on sound biological data.

Because of selenium's propensity to bioaccumulate in aquatic food chains, it is important to carefully assess the *entire* selenium cycle—including downstream transport, transformation, and bioaccumulation—at locations where site-specific criteria are being considered. This is essential because it is possible for two adjacent aquatic systems (e.g., a river and off-channel wetland) to have largely different selenium cycling dynamics. Moreover, the toxicity of selenium to fish can vary depending on time of year (Lemly, 1993a, 1996a). A water quality criterion that is appropriate for one system or time of year may not be sufficient for another. The mechanism of toxicity also demands that the pattern of bioaccumulation be closely scrutinized. For

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example, subtle biochemical dysfunctions can lead to reproductive failure of fish and wildlife; substantial impacts can occur with little or no outward evidence of toxicity (Lemly, 1985a, 1985b).

The degree of mobility (intra- and interhabitat transport), cycling rate and transformation (from selenate to selenite to organic selenium and vice versa), and bioaccumulation of selenium in an aquatic system all influence the toxic threat to biota and, consequently, the adequacy of EPA national guidelines and the need for site-specific criteria. This paper presents a brief overview of the selenium cycle and discusses why site-to-site differences in bioaccumulation and threats to down-gradient aquatic habitats favor the use of a hydrological unit approach for deriving water quality criteria. It also points out the need for EPA to provide an operational framework for use at a local level.

THE SELENIUM CYCLE

Three things can happen to dissolved selenium when it enters an aquatic ecosystem: (1) it can be absorbed or ingested by organisms, (2) it can bind or complex with particulate matter or surficial sediments, or (3) it can remain free in solution. Over time, most of the selenium is either taken up by organisms or bound to particulate matter. Though deposition of biologically incorporated selenium and settling of particulate matter (sedimentation), most of it usually accumulates in the top layer of sediment and detritus. However, because biological, chemical, and physical processes move selenium out of, as well as into, the sediments (Fig. 1), the sediments are only a temporary repository for selenium. Aquatic systems are dynamic, and selenium can be cycled back into the biota and remain at elevated levels for years after waterborne inputs of selenium are stopped (Lemly, 1997a).

Immobilization Processes

Selenium can be removed from solution and sequestered in sediments through the natural processes of chemical and microbial reduction of the selenate form (Se VI) to the selenite form (Se IV), followed by adsorption (binding and complexation) onto clay and the organic carbon phase of particulates, reaction with iron species, and coprecipitation or settling (Fig. 1). Regardless of the route, once selenium is in the sediments, further chemical and microbial reduction may occur, resulting in insoluble organic, mineral, elemental, or adsorbed selenium. Most selenium in animal and plant tissues is eventually deposited as detritus and, over time, isolated through the process of sedimentation. Some

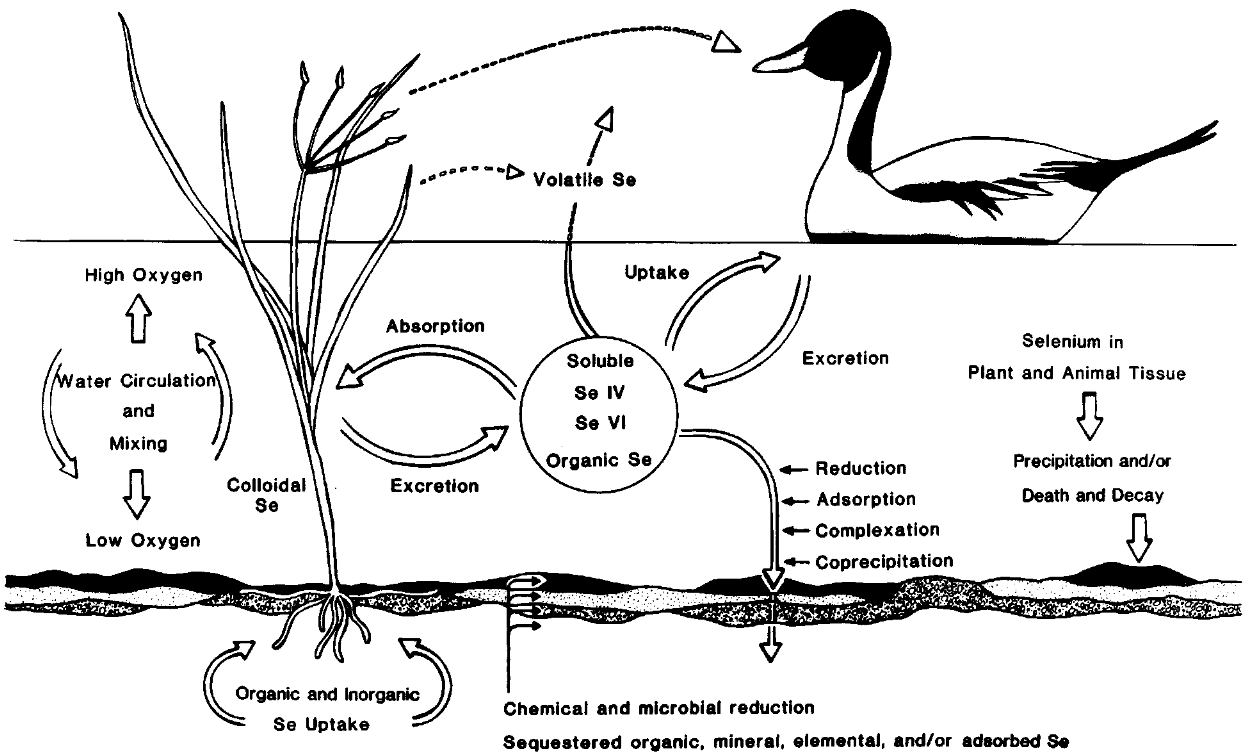

FIG. 1. A highly dynamic system: Biological, chemical, and physical processes cycle selenium into and out of the water, sediments, and biota. Processes that immobilize selenium include chemical and microbial reduction, adsorption, coprecipitation, and deposition of plant and animal tissue; mobilization processes include uptake of selenium by rooted aquatic plants and sediment oxidation due to water circulation and mixing.

selenium, particularly certain organic forms, may be released into the atmosphere through volatilization by chemical or microbial activity in the water and sediments or through direct release by plants.

In total, immobilization processes effectively remove selenium from the soluble pool, especially in slow-moving or still-water habitats and wetlands. Ninety percent of the total selenium in an aquatic system may be in the upper few centimeters of sediment and overlying detritus (Lemly and Smith, 1987).

Mobilization Processes

Selenium in sediments is particularly important to long-term habitat quality because mechanisms present in most aquatic systems effectively mobilize such selenium into food chains and thereby cause long-term dietary exposure of fish and wildlife (Lemly, 1993c, 1997a).

Selenium is made available for biological uptake by four oxidation and methylation processes (Figs. 1–3). The first is the oxidation and methylation of inorganic and organic selenium by plant roots and microorganisms. (Oxidation refers to the conversion of inorganic selenium in the reduced organic, elemental, or selenite forms to the selenite or selenate forms; methylation is the conversion of inorganic or organic selenium to an organic form containing one or more methyl groups, which usually results in a volatile form.) The second process is the biological mixing and associated oxidation of sediments that results from the burrowing of benthic invertebrates and feeding activities of fish and wildlife. The third process is represented by physical perturbation and chemical oxidation associated with water circulation and mixing (current, wind, stratification, precipitation, and upwelling). Finally, sediments may be oxidized by plant photosynthesis.

Two additional pathways provide for direct movement of selenium from sediments into food chains, even when the surface water does not contain the element. These pathways are uptake of selenium by rooted plants and uptake by bottom-dwelling invertebrates and detritus-feeding fish and wildlife. These two pathways may be the most important in the long-term cycling of potentially toxic concentrations of selenium. Thus, rooted plants and the detrital food pathway can continue to be highly contaminated and expose fish and wildlife through dietary routes even though concentrations of selenium in water are low (Lemly and Smith, 1987).

Role of Habitat Variability

The processes regulating selenium cycling are similar in all aquatic habitats, but the relative contribution of each

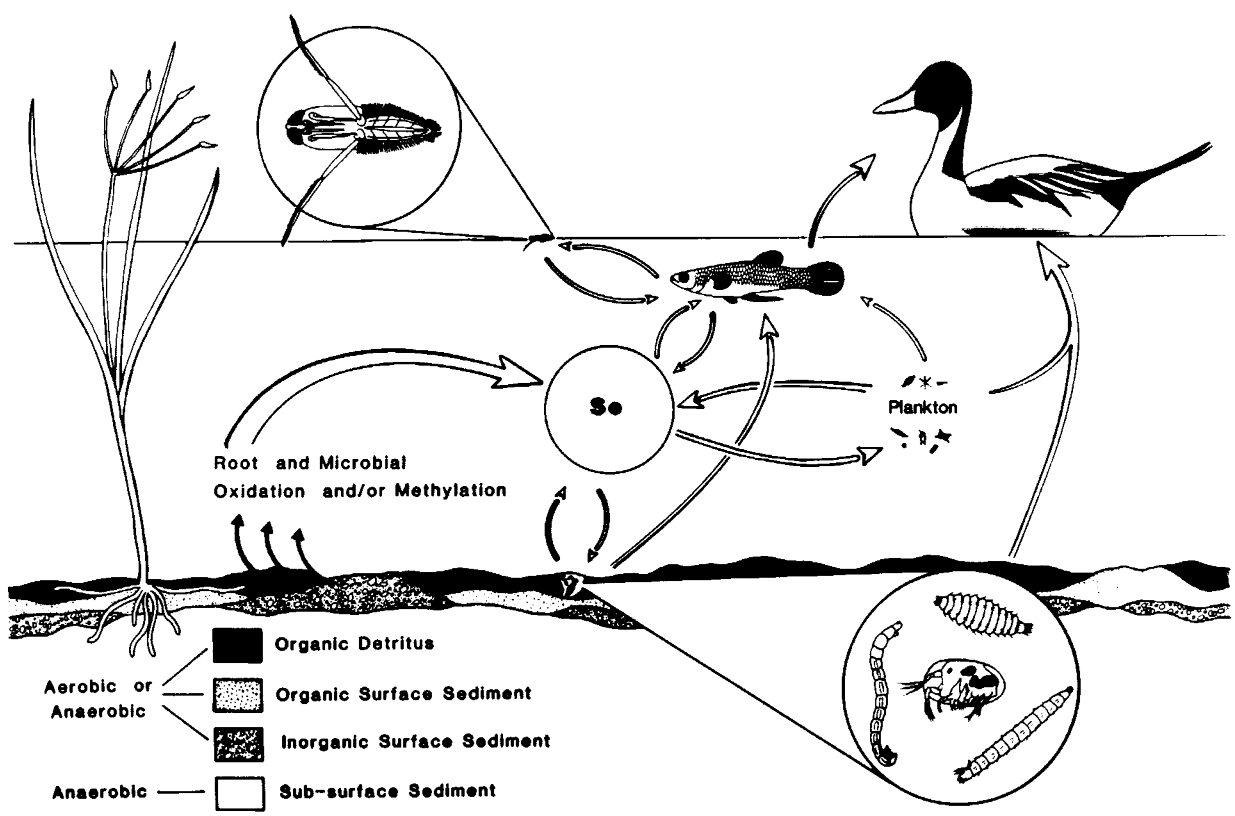

FIG 2. Additional mobilization processes include direct uptake of selenium by benthic invertebrates and oxidation of sediments resulting from plant roots, microorganisms, and the burrowing activity of benthos.

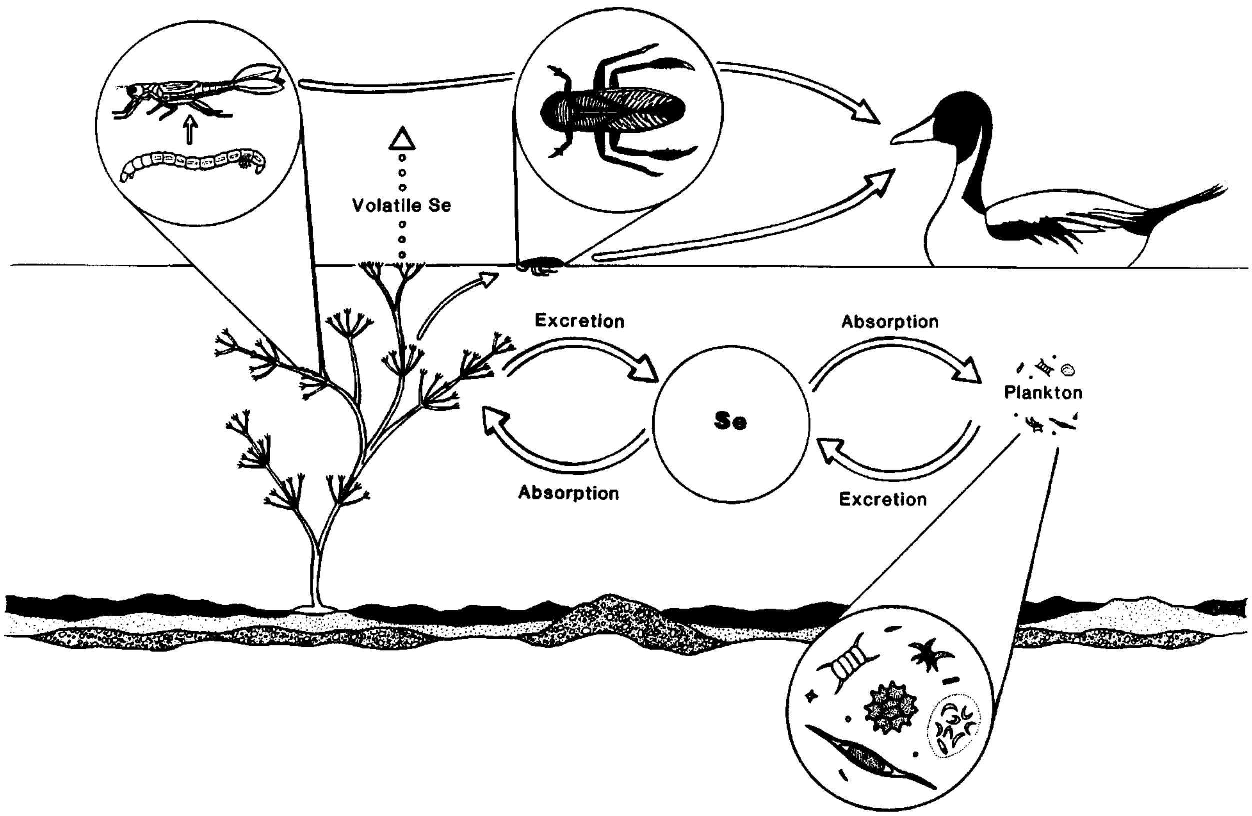

FIG. 3. Dissolved selenium, whether introduced from wastewater discharge or natural geological sources or mobilized from sediments, is readily taken up by aquatic organisms and concentrated in food chains, particularly in wetlands, ponds, and reservoirs. These food pathways converge on top consumer species of fish and wildlife. The effects may be severe even when the concentration of waterborne selenium is low.

process may vary from habitat to habitat. In fast-flowing waters, fine organic sediments such as those produced by the deposition and decay of particulate matter and plant and animal tissue may be rare because they are continually flushed from the system. In such waters, there is little opportunity for a contaminated surface layer of sediment to develop and rooted plants are often scarce. The benthic-detrital components of the system and the associated food pathways thus play a smaller role in the selenium cycle in flowing waters than in slow-water habitats such as wetlands and reservoirs.

The aquatic systems that accumulate selenium most efficiently are shallow, slow-moving waters that have low flushing rates. In these systems, biological productivity is often high and selenium may be trapped through immobilization processes or through direct uptake by organisms. Sediments tend to build up a selenium load that can be remobilized gradually, yet continually, through detrital and planktonic food. These habitats are also some of the most important feeding and breeding habitats for fish and wildlife, especially waterfowl and shorebirds.

Several habitat types often occur together in one aquatic system. For example, rivers may have fast-flowing waters, slow-moving pools, and standing backwater areas, all within a few hundred meters. The degree of fish and wildlife exposure to selenium varies among habitats according to intensity of use, type of use, and relative contributions of the various processes that regulate selenium cycling.

Water quality criteria for selenium should be based on an assessment of the degree of contamination and toxicological risk which, in turn, will depend on the spatial and temporal variation of the selenium cycle at the site under consideration. In addition to protecting resident biota at the "site," it is important that criteria protect against possible impacts to down-gradient aquatic habitats as well. For example, a criterion that is appropriate for a stream or river where low bioaccumulation occurs may result in *seemingly* harmless concentrations of selenium becoming a problem in downstream impoundments or in off-channel bays and wetlands where bioaccumulation is greater (Lemly, 1998) (Fig. 4).

IMPORTANCE OF BIOACCUMULATION

It is critical for those who develop water quality criteria for selenium to incorporate the bioaccumulation phenomenon into the derivation process. The major principle to

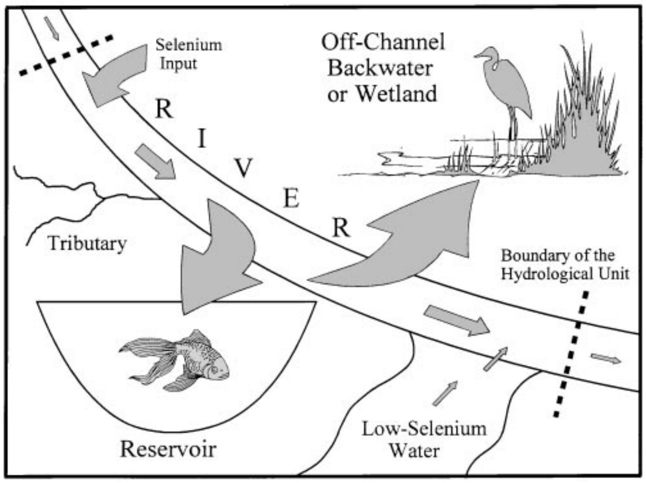

FIG. 4. Illustration of the "hydrological unit" concept. Arrows indicate the relative concentrations of selenium. The interconnected parts of a unit may include several types of aquatic habitats, for example, a main-stem river, a wetland, and a reservoir. Given equal waterborne concentrations of selenium, the degree of bioaccumulation and toxicity to wildlife can be substantially greater in off-channel lentic systems than in a river or stream. It is necessary to guard against downstream transport of hazardous concentrations of selenium that could result if a relaxed or liberal criterion were set for the river. Establishing a single criterion for the entire hydrological unit is the best way to ensure protection of fish and wildlife.

remember is that reproductive effects in fish and aquatic birds are the most sensitive biological indicators of aquatic ecosystem-level impacts of selenium. Selenium in water can be concentrated from 100 to more than 30,000 times in the food organisms eaten by fish and wildlife, which exposes them to a highly concentrated dietary source of contamination. Biomagnification may also occur, resulting in a two- to sixfold increase in selenium between primary producers and forage fish. Moreover, if the ecosystem is allowed to reach equilibrium such that recycling of selenium from sediment occurs, the detrital food pathway can deliver toxic doses of selenium for many years even if waterborne sources are eliminated (Lemly 1982, 1985a, 1997a).

A significant portion of the selenium consumed by wildlife is passed to their offspring in eggs, where it can kill developing embryos outright or induce a variety of lethal or sublethal teratogenic deformities (Lemly, 1993c). However, parents can consume a selenium-laden diet and experience partial or complete reproductive failure without exhibiting symptoms of selenium toxicosis themselves. Moreover, aquatic food organisms of wildlife strongly bioaccumulate selenium—hundreds to thousands of times the waterborne concentration—but are unaffected by tissue residues that are high enough to cause reproductive failure when consumed by fish and aquatic birds. Thus, bioaccumulation in aquatic food chains and dietary transfer to eggs cause otherwise innocuous concentrations of waterborne selenium to become toxic (Lemly, 1993b). Establishing water criteria that prevent this amount of bioaccumulation in aquatic food chains should be the goal of site-specific derivations.

The degree of selenium accumulation in major ecosystem components (sediments, invertebrates, fish, birds) can be used to precisely evaluate local conditions and determine if existing or proposed criteria are appropriate, too conservative, or too liberal. This process can be applied consistently and uniformly, regardless of location or habitat type, i.e., in wetlands (seasonal or permanent), streams, rivers, lakes, reservoirs, and other impoundments. Criteria that prevent bioaccumulation from reaching levels sufficient to cause dietary toxicity and reproductive effects in fish and wildlife will afford protection to other aquatic life as well.

GATHERING AND USING SITE-SPECIFIC INFORMATION

The first step in establishing appropriate criteria is to obtain information on selenium residues and associated biological effects. This is done to provide an empirical foundation for evaluating a criterion, which may be national, state, or local, and which may be currently in place or under consideration. Some of the tools available for this process include a hazard assessment protocol (Lemly, 1995b, 1996b, 1997b), a teratogenic deformity index for fish (Lemly, 1997c), and risk thresholds for waterbirds (Skorupa and Ohlendorf, 1991). Guidance for measuring selenium residues and interpreting tissue concentrations and biological effects thresholds is also available (Lemly, 1993b, 1998; Skorupa *et al*., 1996). Collectively, these tools make it possible to examine a criterion value in the context of site-specific selenium concentrations and observed potential toxicity to fish and aquatic birds, including threats to down-gradient aquatic habitats. Several outcomes and conclusions are possible: a criterion may be appropriate as it is, or it may be inappropriate and need to be raised or lowered. If the empirical evidence indicates that revision is necessary, a simple procedure that uses the degree of bioaccumulation and magnitude of toxic effects can be followed to establish the new criterion (Lemly, 1998).

SETTING CRITERIA FOR HYDROLOGICAL UNITS

Perhaps the most important point conveyed in this paper is that the physical area from which measurements are taken to evaluate selenium residues and biological effects, i.e., the database for setting site-specific criteria, must encompass more than an isolated segment of river, a tributary stream, etc. The overarching principle for establishing environmentally sound water quality criteria for selenium is that bioaccumulation be held below levels that pose significant threats to biota. For this to be accomplished, hazards to down-gradient aquatic habitats, most or all of which may fall outside the "site" under consideration, must be assessed and accounted for.

Differences in selenium cycling between sites is one of the main reasons that EPA national criteria are coming into question more frequently—one size does not fit all. However, the reasoning that individual sites need specific criteria can be taken too far. For example, isolating a segment of a river and setting a criterion for it when biota in the receiving waters downstream may suffer is not a prudent approach. Because of hydrological connections between the various aquatic habitats that may be present in a watershed basin—wetlands, rivers, streams, and impoundments—ecological risk and toxic threat from selenium contamination are also connected.

The hydrologically connected parts of a basin downstream of a selenium discharge (natural or synthetic selenium source), extending to the point at which new sources of low-selenium water dominate the hydrology (e.g., confluence with larger tributary or river, spring or groundwater inflow), should be the area evaluated and given a specific criterion, not isolated components (Fig. 4). Thus, a hydrological unit should be identified and used as the "site" for the purpose of setting criteria.

Designating hydrological units is a departure from the traditional concept of what constitutes a "site." Consequently, it may be more useful to refer to criteria as watershed specific or basin specific rather than site specific. While still providing for focused, local refinement of EPA national criteria, this method ensures an integrated assessment of ecosystems within a watershed and allows basinwide protection of aquatic life. Importantly, criteria derived in such a fashion will reflect the transport and cycling of selenium within the entire hydrological unit rather than simply focusing on a small, artificially designated segment of the system, i.e., a section of river, a tributary stream, etc.

Failure to use a hydrological unit approach can set the stage for significant biological and legal problems. Consider, for example, the following scenario. A chronic exposure criterion of 20 μg/liter is derived and adopted for a 10-km segment of river immediately downstream of a municipal wastewater treatment plant based on the finding that there are no toxic impacts and little bioaccumulation. A few kilometers downstream of this river segment there is a 250-ha off-channel wetland that is used by wildlife for feeding, spawning, and nesting. Part of the wetland is a state wildlife management area and part is under private ownership. The 20 μg/liter selenium that is permitted in the river flows into the wetland, bioaccumulates in aquatic food chains, and causes toxic impacts to fish and bird embryos. Several questions arise. What is the appropriate criterion for the wetland? Should it be imposed on the river where no problems are occurring? Is it feasible to set two criteria—one for the river and one for the wetland—when the two are hydrologically connected? Who is liable for toxicity to wildlife and what recourse is possible? Will litigation be necessary to resolve the dilemma and, if so, will EPA be one of the litigants?

This hypothetical case is becoming reality in several Western states as natural resource management agencies, local municipalities, industry, and private landowners become aware of the threat selenium poses to fish and wildlife and understand the liabilities that ensue if poisoning occurs (e.g., Guglielmone, 1995; Margolin, 1979). The task of settling "site-specific" criteria issues may involve complex questions of land and water jurisdiction if the hydrological unit affected by selenium includes several municipalities and counties or crosses state boundaries.

CONCLUSIONS AND RECOMMENDATIONS

Selenium contamination of aquatic ecosystems is a growing environmental concern at both local and national levels in the United States. It is important that those seeking to develop site-specific water quality criteria for selenium have clear guidance on how to go about it. The guidance should be in the form of an instructional framework for designating hydrological units, obtaining and interpreting information on selenium residues and biological effects, and prescribing actions to be taken in setting local criteria. The framework should lay out a clear step-by-step process that will leave no doubt as to the information required and the process involved. The present paper and its supporting references can be the basis for a framework document that provides appropriate guidance to states and municipalities.

Logically, EPA should develop and provide the framework since it is responsible for reviewing information and approving site-specific modifications of national criteria. However, the Agency has not been proactive in this regard and continues to wrestle with each local selenium problem as it arises. Without such a framework the Agency will likely be drawn into litigation on a case-by-case basis as the national criteria are challenged (which is already occurring in some Western states). Litigation should be the last resort because it generates distrust for EPA by those being regulated, and could seriously tarnish the Agency's credibility as a leader in environmental protection, i.e., by promoting the view that the Agency is not providing constructive assistance on selenium issues at a local level and does not get involved unless threatened by a lawsuit.

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